Lecture 16. Light scattering and absorption by atmospheric particulate. Part 4: Scattering and absorption by non-spherical particles: DDA and FDDA methods. Examples.

Objectives:

1. Basics physics of dipole interactions.

2. Outline of the DDA method.

3. Effects of particle nonsphericity on main aerosol optical characteristics.

Recommended Reading:

Stephens G. Remote sensing of the lower atmosphere. 1994.

Chapter 5.2

1. Basics physics of dipole interactions.

Recall Lecture13: molecular scattering can be described by considering a molecule as the ***single isolated dipole*** (Rayleigh limit: x << 1)

In the Rayleigh limit, perpendicular and parallel components of scattered intensities in the far-field are given by Eq.[13.22]

$$I_r = I_{0r} k^4 \alpha^2 / r^2$$

$$I_l = I_{0l} k^4 \alpha^2 \cos^2(\Theta) / r^2$$

where α is the polarizability of the particle and it relates to the refractive index via Lorentz-Lorentz formula (also known as Clausius-Mossotti formula) as (Eq.[13.28])

$$\alpha = \frac{3}{4\pi N_s}\left(\frac{m^2 - 1}{m^2 + 2}\right)$$

Rayleigh-Gans limit is $x(m-1) \ll 1$ ($x \ll 1$ is not required). In Rayleigh-Gans limit, the field inside the particle is the same as the incident. The field is not reflected by the particle and there is no phase difference across particle.

Conceptual model: A particle may be divided into "dipoles" of size $2\pi d/\lambda < 1$.

EM field of two isolated dipoles:

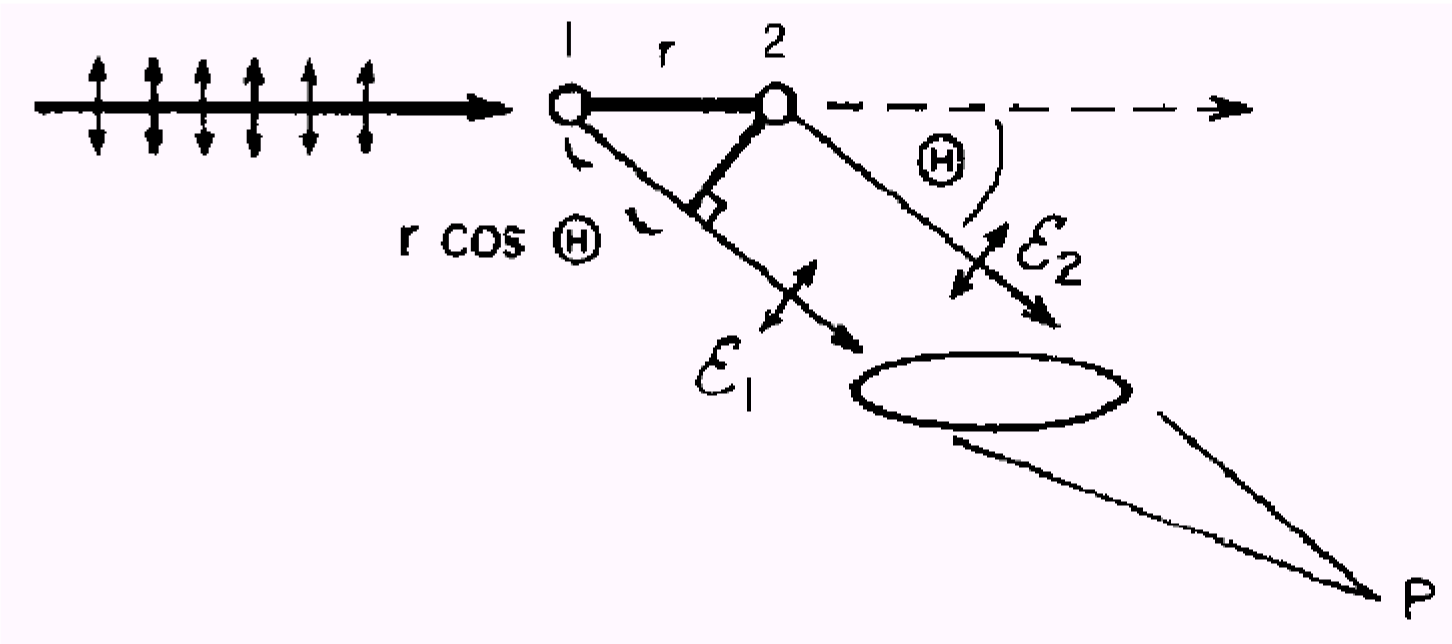


Figure 16.1 Two isolated dipoles scatter the incident EM field into all directions. (Stephens, 1994). An observer at point P will measure the superposition of scattered waves of two dipoles, propagating into the direction of observation (i.e., in scattering angle Θ). The interference of these two waves depends on the phase difference caused by the relative path difference.

The phase difference between two scattered waves at point P is the difference in path length of two waves

$$\Delta\delta = x(1 - \cos\Theta) \qquad [16.1]$$

where x is the size parameter. Thus the scattered field is

$$E_{1+2} \sim E_1 \exp(-i\delta) + E_2 \exp(-i(\delta + \Delta\delta)) \qquad [16.2]$$

and the detected intensity (averaged over the full cycle)

$$I_{1+2} \sim E_1^2 + E_2^2 + 2E_1E_2\cos(\Delta\delta) \qquad [16.3]$$

If $\boldsymbol{E_1}=\boldsymbol{E_2}$

$\Delta\delta = \pi, 3\pi, 5\pi \ldots$ => fields cancel (out of phase)

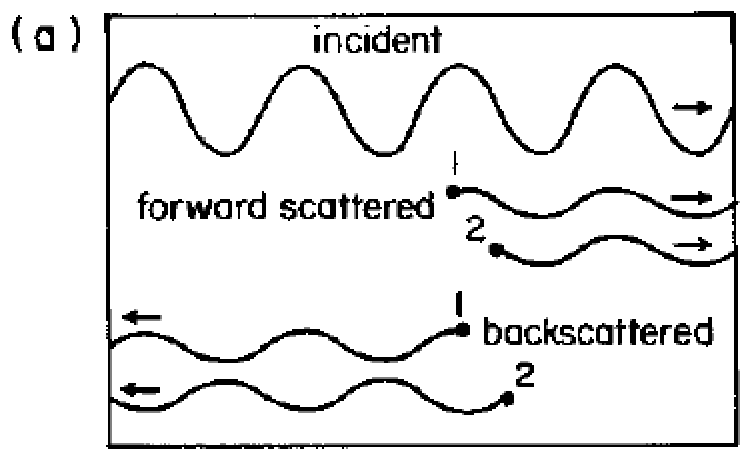
$\Delta\delta = 0, 2\pi, 4\pi, \ldots$ => fields reinforce (in phase)

NOTE: The forward scattering waves are always in phase.

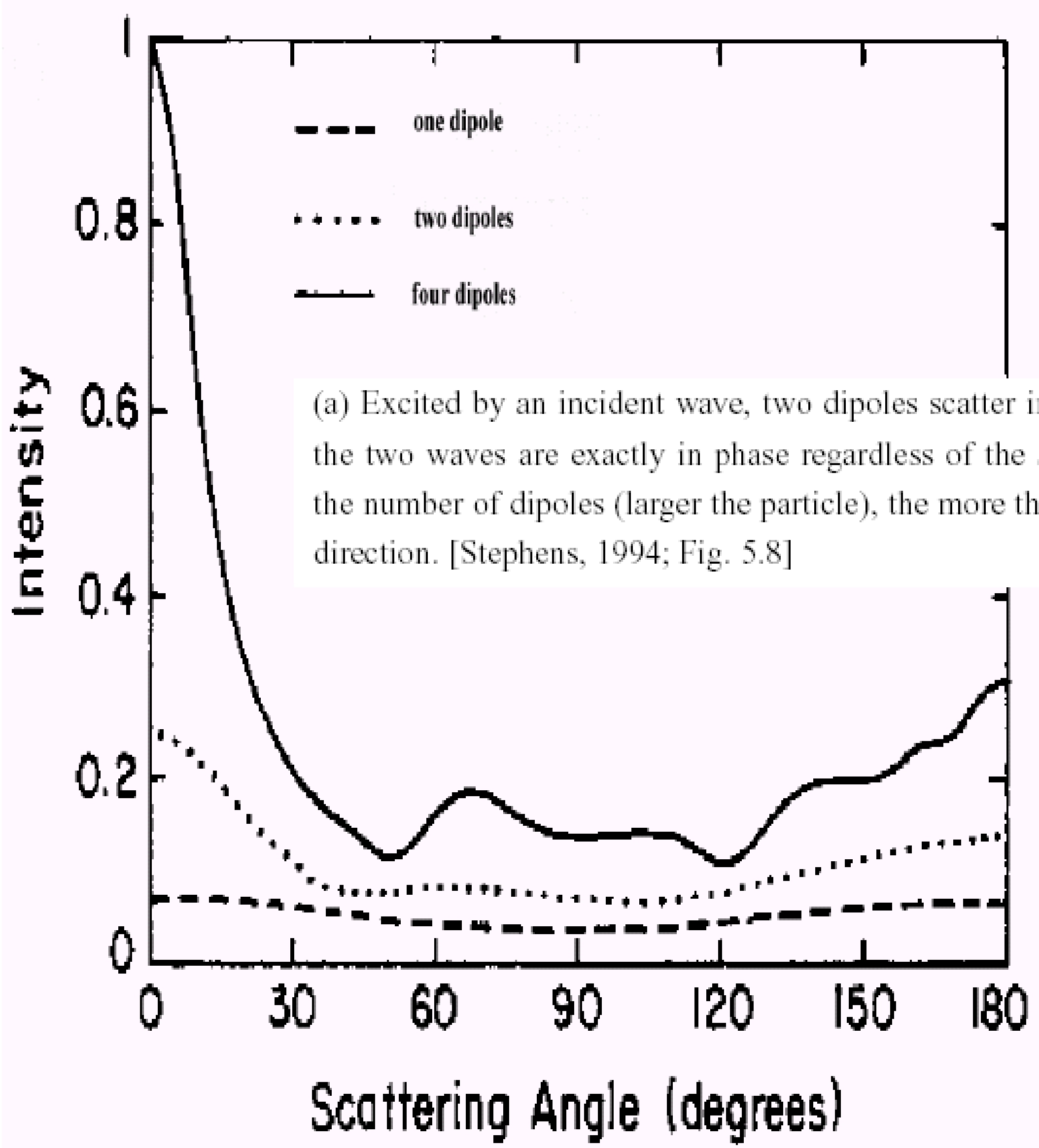
- In general case for nonspherical particle represented by many dipoles, the phase difference depends on both the distance between dipoles and the direction of scattering (except for $\Theta = 0^0$, i.e. forward scattering) => the scattered radiation is a complex superposition of individual scattered waves of many different phase differences.

Scattered waves from dipoles are in phase for forward direction $\Theta = 0$, giving constructive interference → forward peak in phase function.

Destructive and constructive interference alternates as Θ increases → oscillations in phase function. More oscillations for larger particle.



- The larger the particle, the higher intensity scattered in the forward direction and the greater the forward to backward asymmetry.



(a) Excited by an incident wave, two dipoles scatter in all directions. In the forwarrd direction, the two waves are exactly in phase regardless of the separation of the dipoles. (b) The greater the number of dipoles (larger the particle), the more they collectively scatter towards the forward direction. [Stephens, 1994; Fig. 5.8]

Figure 16.2 Intensity of dipoles lied on one line at the distance of one wavelength and interacted with each other (Bohren, 1987).

- In addition to phase differences, the scattered field is affected by interaction of dipoles with each other.

Consider a particle composed of many dipoles. The scattered field is incident field plus the fields produced by each dipole

$$E_{sc} = E_{inc} + \sum E_{dipoles} \tag{16.4}$$

The dipole moment of j-th dipole is

$$p_j = \alpha_j E_{dipole,j} \tag{16.5}$$

where α_j is the polarizability of the dipole and $E_{dipole,j}$ is he field acting on the dipole which is the superposition of incident field and the fields caused by other dipoles. Thus

$$p_j = \alpha_j [E_{inc,j} - \sum_{j \neq k} A_{jk} p_k] \tag{16.6}$$

where $-\sum_{j \neq k} A_{jk} p_k$ is the contribution from the electric field at j-th dipole from the k-th dipole.

NOTE: Solving Eqs.[16.4]-[16.6] for all p_j lies the basis of the Discrete Dipole Approximation (DDA) method.

2. Outline of the DDA method.

DDA (Discrete Dipole Approximation) method enables computation of optical properties of arbitrary shaped, inhomogeneous, and anisotropic particles.

A numerical method for scattering from any shape particle that is not too large ($x < 5$). See review paper by Draine and Flatau, J. Opt. Soc. Am. A, **11,** 1491.

NOTE: DDSCAT is a FORTRAN implementation of the DDA technique. The code and user guide are openly available at http://www.astro.princeton.edu/~draine/DDSCAT.html

Basic principles: In DDA, the particle is replaced by an array of polarizable points (dipoles), and then the electromagnetic scattering problem for an incident periodic wave interacting with this array of point dipoles is solved exactly.

For a particular incident E field, the linear system may be solved for the dipole moments $\mathbf{p}_k$.
The far field scattering properties are calculated from the $\mathbf{p}_k$.

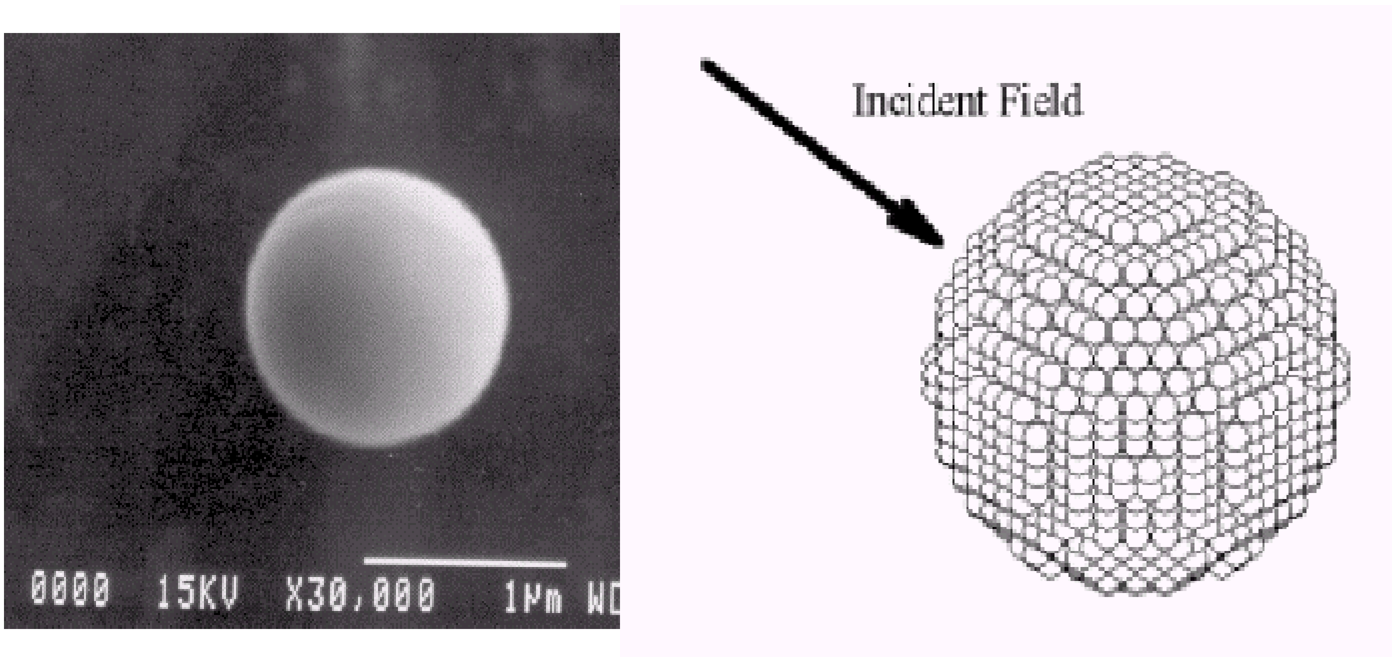
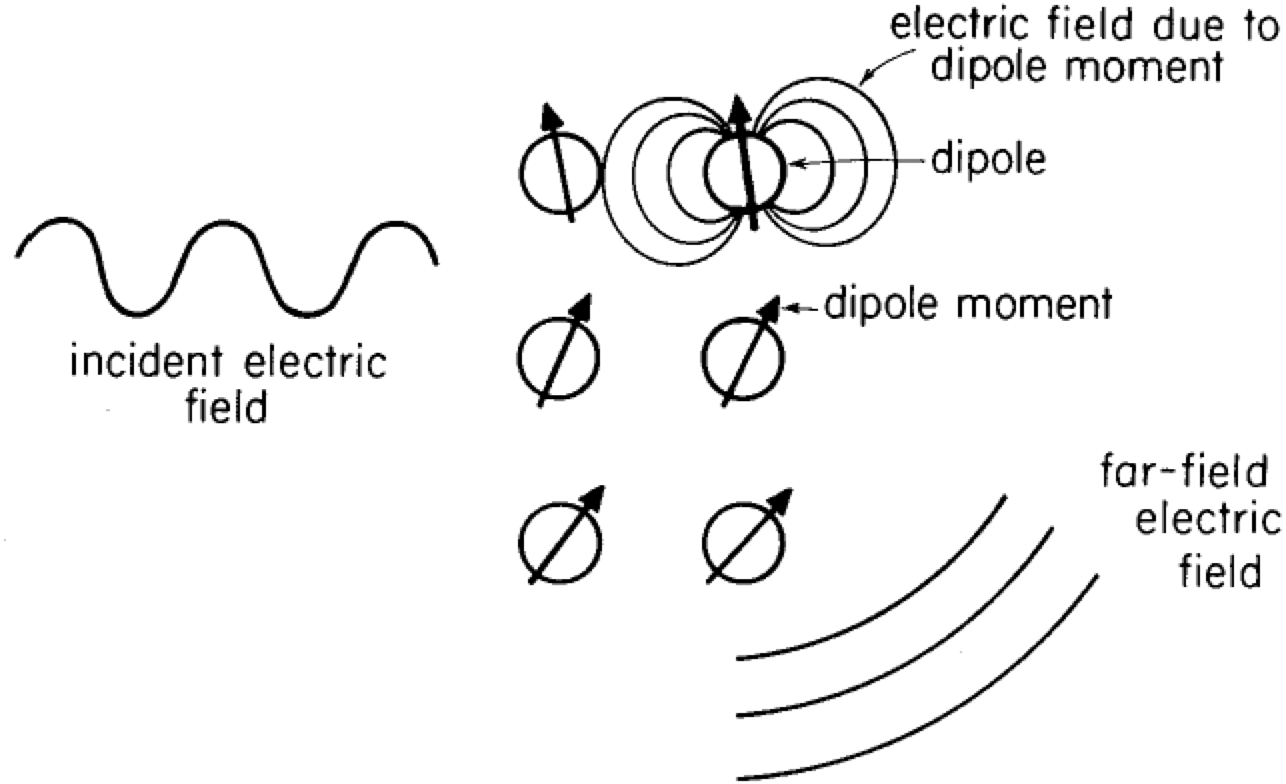


Figure 16.3 A spherical particle represented by individual dipoles.



The Discrete Dipole Approximation divides a particle up into many dipoles which are small compared to the wavelength. The oscillating electric field produced by each dipole depends on the dipole moment. The dipole moment depends on the applied electric field from the incident field and other dipoles and the dielectric properties of the dipole. The far-field electric field is a sum of the dipole fields from all the dipoles.

Advantages: DDA can be applied to particles having any shape and composition (i.e., homogeneous or aggregates)

Applicability and limitations of DDA:

DDA is completely flexible regarding the geometry of a particle, being only limited by the need to use an interdipole distance ***d*** small to satisfy

$$\frac{2\pi}{\lambda}|m|d < 1 \qquad [16.7]$$

where m is the complex refractive index of the particle.

If a particle of volume V is represented by an array of N dipoles, located on a cubic lattice with lattice spacing ***d***, then

$$V = Nd^3 \qquad [16.8]$$

The size of the particle can be characterized by the "effective radius" $\boldsymbol{a_{eff}}$ as

$$a_{eff} = (3V/4\pi)^{1/3} \qquad [16.9]$$

i.e., $\boldsymbol{a_{eff}}$ is the radius of an equal volume sphere.

Then the size parameter is $x = \frac{2\pi}{\lambda} a_{eff}$

and it can be related to N as

$$x = \frac{2\pi}{\lambda} a_{eff} = \frac{62.04}{|m|} \left(\frac{N}{10^6} \right)^{1/3} |m| \frac{2\pi}{\lambda} d \qquad [16.10]$$

The number of dipoles N ~ a_{eff}^{3}, so as particle size increases, the very large number of dipoles are required. Therefore, DDA is limited by the size parameter of about 15 (or the number of dipole is up to N=10^6).

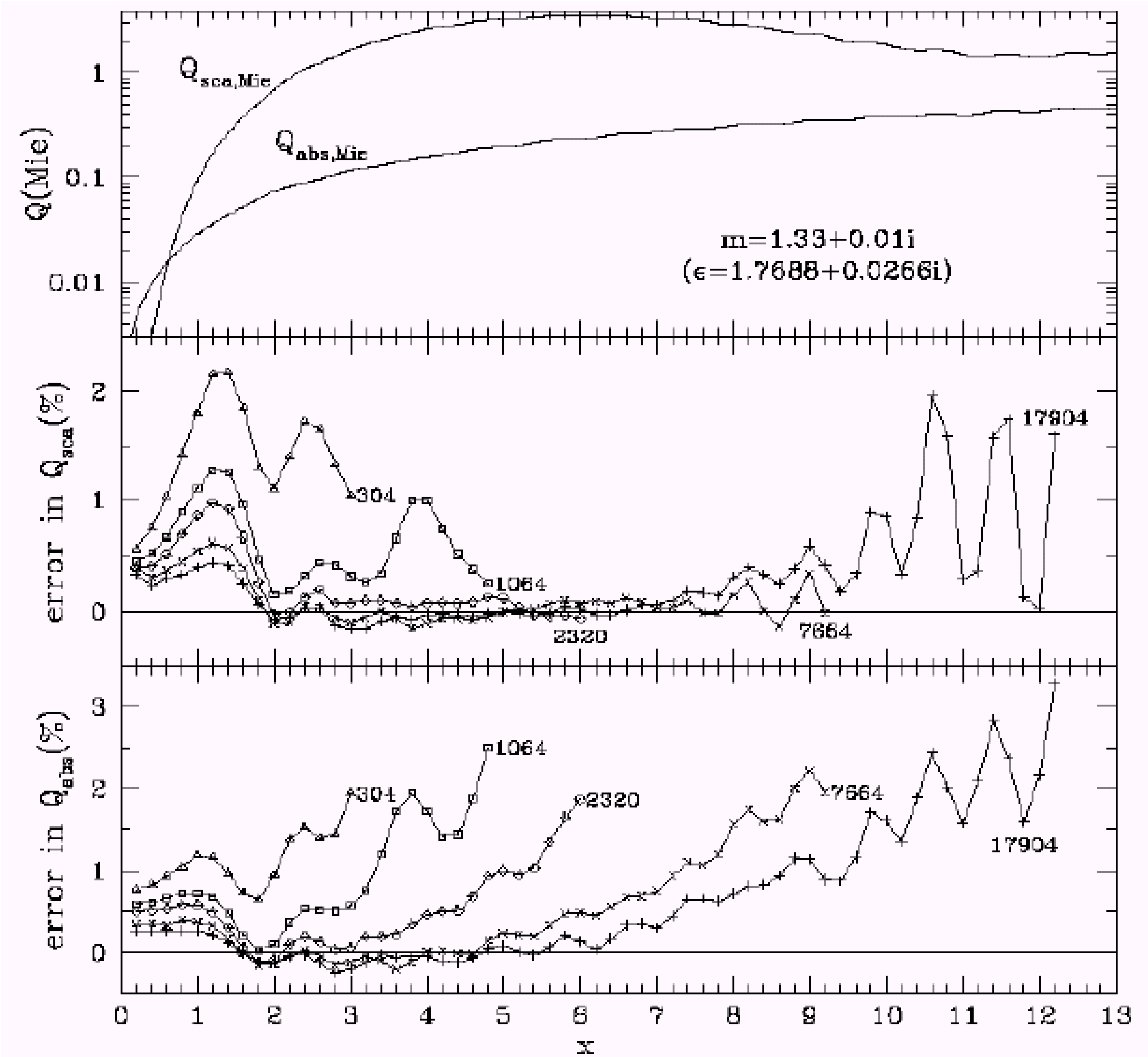


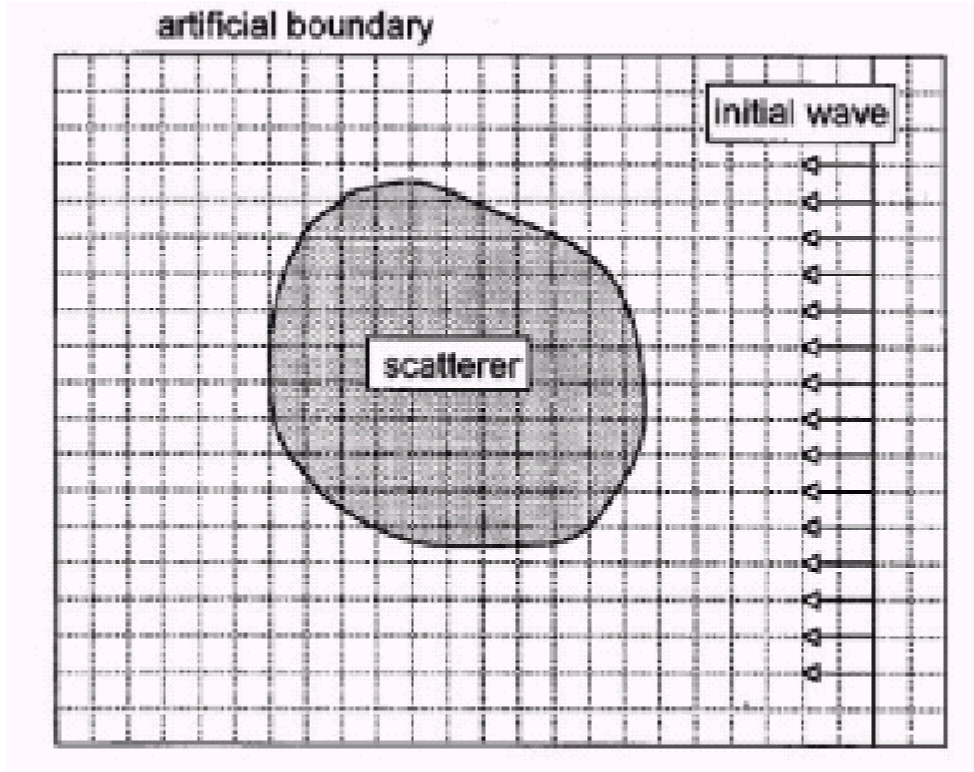
Fig. 16.4 Scattering and absorption efficiencies for a sphere with m=1.33+0.01i.

The upper panel shows exact calculation from Mie, whereas the middle and lower panels show fractional errors in Q_s and Q_e, calculated with DDSCAT for different numbers of dipoles N (Draine and Flatau, 1994)

3. Outline of the FDTD method.

Finite Difference Time Domain, FDTD, method enables calculations of optical properties of particles of complicated geometries and compositions.

Basic principles: FDTD solves the Maxwell's curl equations (first two equations in eq.[14.1]) in the time-domain by introducing a finite difference analog. The space containing a scattering particle is dicretized by using a grid mesh. The existence of the particle is represented by assigning suitable electromagnetic constants in terms of permittivity, permeability and conductivity (depending on particle properties) over the grid points.



Advantages: FDTD can be applied to particles having any shape and composition.

Limitations: Known implementation problems (for instance, staircasing effect - due to selection of Cartesian mesh grid)

Limited range of size parameters (up to x =15-20)

➢ **Applications**

Ice crystals:

Yang and Liou: FDTD for size parameter ~15 and ray-tracing (for x >15)

Dust particles:

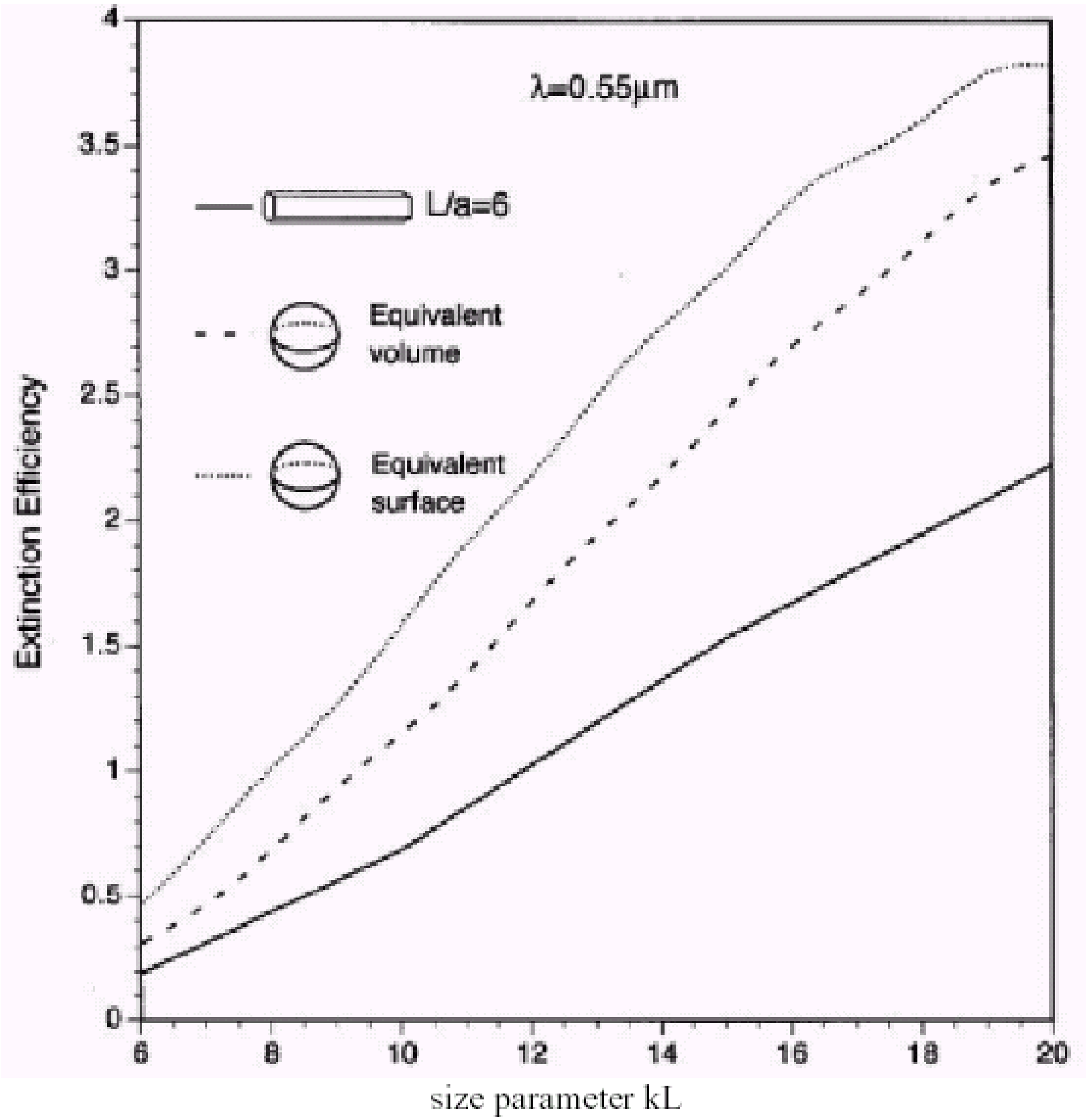
Mishchenko et al.: T-matrix applied to a mixture of ellipsoids

Kalashnikova and Sokolik (2002): DDA applied to SEM data of dust particles (see Lecture 16)

Soot aerosol:

Mackowski et al.: modified T-matrix applied to fractal-like sphere clusters.

How to define an equivalent sphere:



4. Effects of particle non-sphericity on main aerosol optical characteristics.

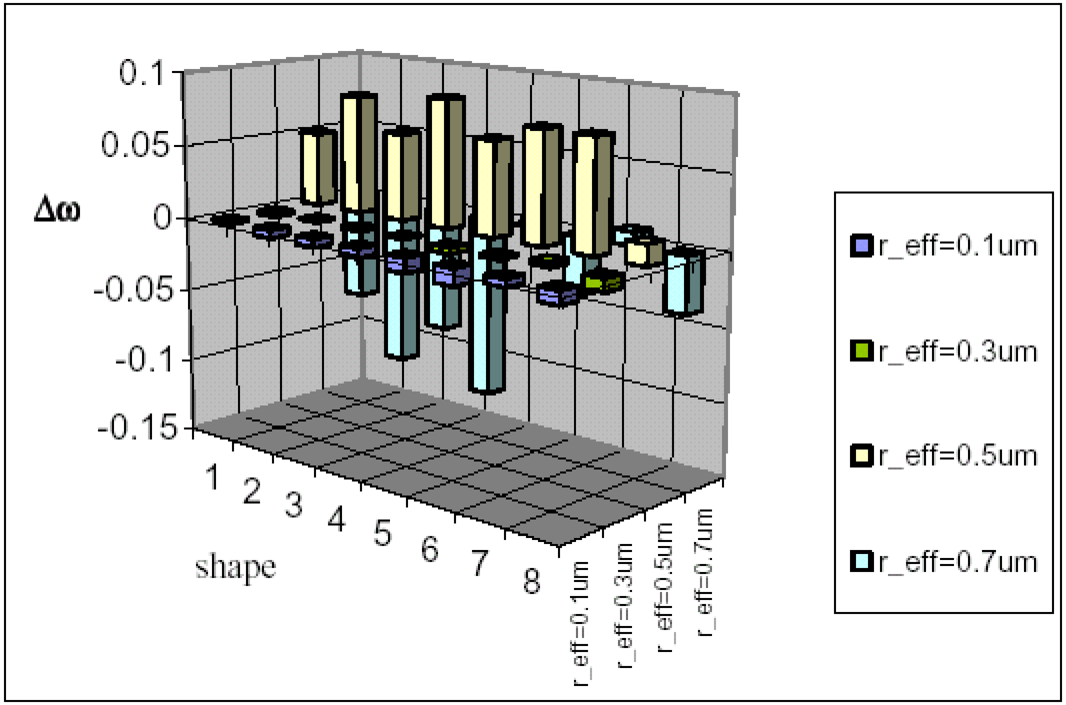
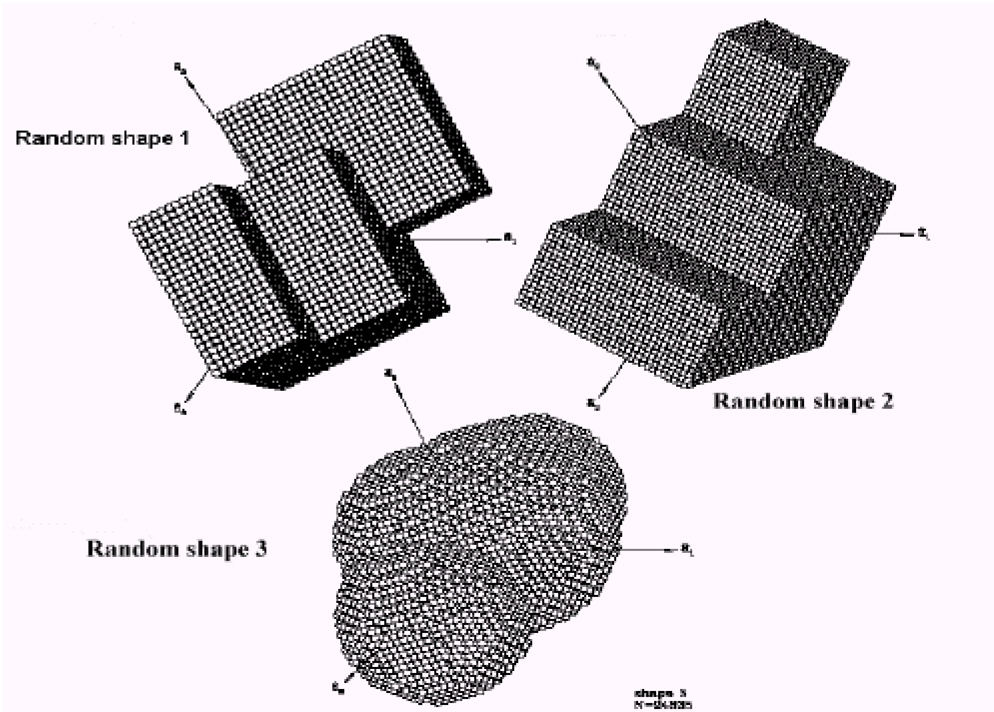


Figure 16.5 Differences in **the single scattering albedo** between spherical and nonspherical particles of equal volume calculated with DDA. Nonspherical shapes: (1) ellipsoid, (2)cylinder, (3) hexagon, (4) rectangular, (5) tetrahedron, (6) –(8) random 1, 2 and 3, respectively. (From Kalashnikova and Sokolik, 2002).



- Compare to a sphere, the nonspherical particle of same composition and volume can result in lower or higher values of the single scattering albedo.

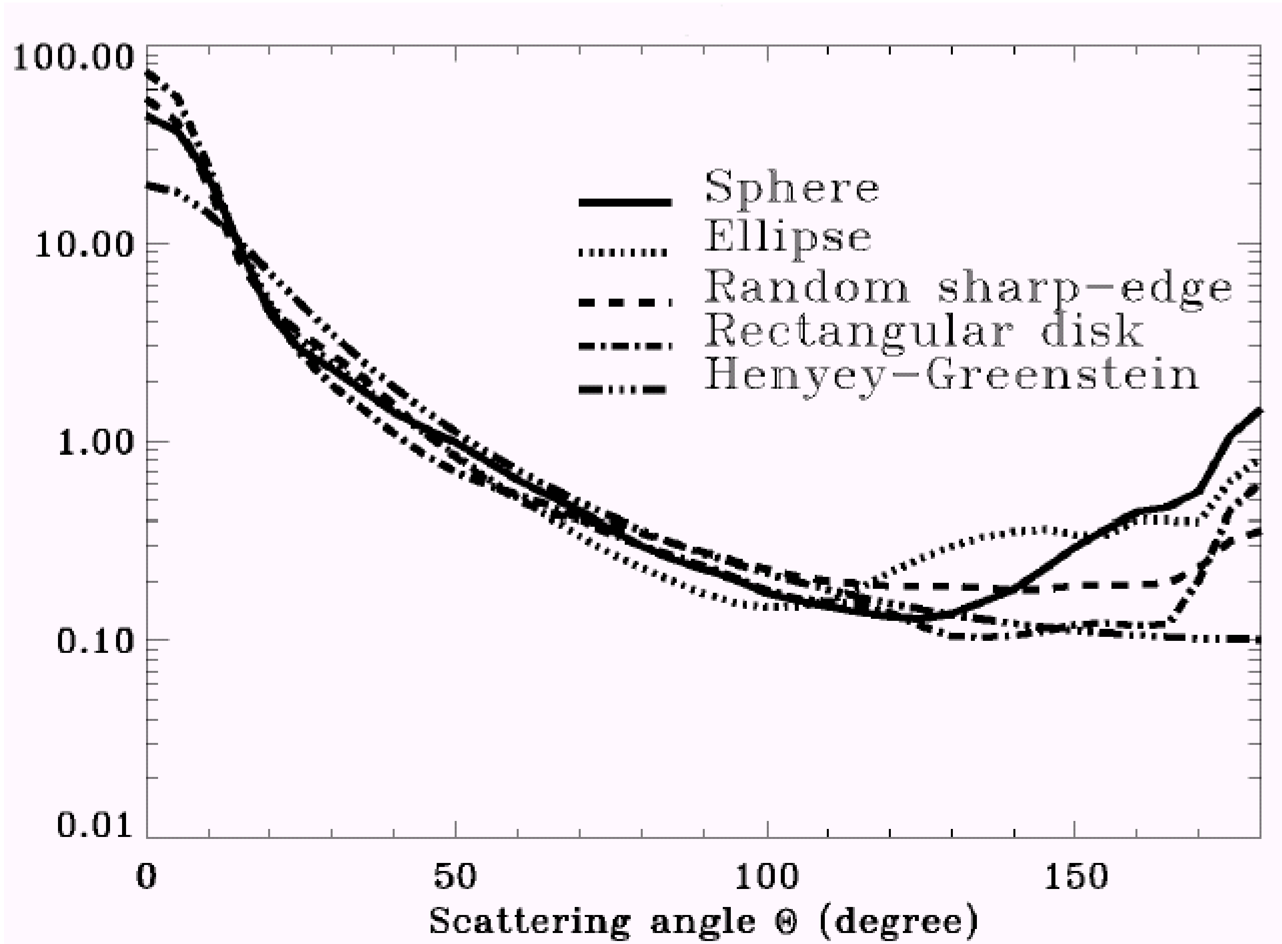


Figure 16.5 Scattering phase function calculated with DDA for a log-normal size distribution with r_0= 0.5 μm, at λ = 0.5 μm , m = 1.51 +i0.002 (From Kalashnikova and Sokolik, 2002).

- Nonspherical particles cause lower scattering in the backscattering directions but larger forward scattering relative to equal volume spheres of the same composition.

Henyey-Greenstein scattering phase function is often used in radiative transfer calculations and is defined as

$$P_{HG}(\Theta) = \frac{1 - g^2}{(1 + g^2 - 2g\cos\Theta)^{3/2}} \qquad [16.11]$$

where g is the asymmetry parameter.

NOTE: Henyey-Greenstein scattering phase function result in lower forward and backward scattering relative to the scattering phase function of spherical and nonspherical shapes.